PLANT APPLICATION OF NUCLEAR
CRITICALITY PREVENTION PRINCIPLES

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INTRODUCTION

The prevention of an accidental, uncontrolled nuclear chain reaction, or nuclear excursion, is of paramount concern in the design and operation of a nuclear fuels processing plant. The probability of a nuclear excursion is much less than for a chemical explosion or fire in a well-designed plant. One might add, with some justification, that in a well-designed plant, the consequences of a nuclear excursion are much less than the consequences of an explosion or an uncontrolled release of hazardous chemicals. Problems associated specifically with a nuclear excursion, however, include the resulting radiation hazard and the potential for a spontaneous recurrence, as well as the public relations aspects. These factors could cause an extensive curtailment in processing operations.

With the above considerations as a background, this paper discusses the application, in both process and facility design and in plant process and operational control, of some of the more important principles of nuclear criticality safety. Included in the discussion are the definition of and conditions affecting criticality; criticality prevention methods, philosophy, and control bases; and finally, applications of criticality prevention principles and philosophy in flowsheet and facility design and in plant operation.

REQUIREMENTS AND POLICY

The application of criticality prevention principles to the processing operations in the Hanford Purex Plant is governed by the AEC requirements for criticality safety which are, in turn, translated into a formal company policy guide and operating instruction. The policy guide and operating instruction require, in essence, that:

- a. A formal system of criticality prevention specifications shall be in effect for any facility or operation in which fissile material is handled in significant quantity.
- b. The specifications shall be implemented with written administrative procedures.
- c. The actions of each organization involved with fissile materials shall be audited periodically by another knowledgeable group for compliance with established criteria and specifications.

The preparation of criticality prevention specifications is preceded by a comprehensive analysis of the criticality safety aspects of the facility. Each equipment piece and each processing step is reviewed for the possibility of criticality; the principle on which nuclear safety will depend is identified and a rationale prepared demonstrating that criticality cannot occur or that the probability is very low. Criticality prevention specifications defining the limits or restrictions necessary to maintain the probability of criticality at the lowest practicable level are then prepared based on the findings of the safety analysis.

DEFINITION AND CONDITIONS FOR A NUCLEAR EXCURSION

Before considering the principles and methods of nuclear criticality prevention, it may be helpful to review the conditions required for a nuclear excursion to occur. First, what is a nuclear

excursion or a criticality incident? A nuclear excursion in a chemical processing plant has been referred to, somewhat irreverently, as an unscheduled homogeneous reactor experiment. This definition, however, does not clarify the question. Clayton⁽¹⁾ has defined the point of criticality as *a self-sustaining nuclear chain reaction in which there is an exact balance between the production of neutrons [from spontaneous fission] and the loss of neutrons in the absence of extraneous neutron sources*. Another way of expressing this relationship is that the ratio of neutrons in one generation to the number of corresponding neutrons in the preceding generation (the reproduction factor) is equal to unity. When the neutron loss from a system, either through leakage or by absorption in nonfissile material, exceeds the neutron production rate, the system is subcritical and a self-sustaining chain reaction will not be possible. If, on the other hand, the neutron loss is less than the neutron production rate, a divergent chain reaction or nuclear excursion will occur.

CRITICALITY PREVENTION METHODS

A brief review of the conditions required for a critically safe system should, perhaps, lead one to envision numerous methods for assuring that a system remains subcritical. Acceptable methods for maintaining nuclear safety at the Hanford Purex Plant have been defined by Tomlinson⁽²⁾ and are listed in Table I and discussed below in order of decreasing safety assurance.

(1) HW-71666, November, 1961, E. D. Clayton and W. A. Reardon, *Nuclear Safety and Criticality of Plutonium*

(2) ARH-468, April 2, 1968, R. E. Tomlinson, *Technical Criteria for the Prevention of Criticality-Chemical Processing Division*

TABLE I
CRITICALITY PREVENTION METHODS

1. GEOMETRICALLY
SAFE
EQUIPMENT	◦ Subcritical under all possible conditions (fully reflected metal) ◦ For ^{239}Pu: Cylinder - 1.7 inches in diameter
Slab - 0.25 inches thick
2. GEOMETRICALLY
FAVORABLE
EQUIPMENT	◦ Subcritical under worst foreseeable conditions of reflection and concentration. ◦ Critical Dimension $\leq 85\%$ of minimum calculated value ◦ For $\text{Pu}(\text{NO}_3)_4$, 3 wt. % ^{240}Pu, Cylinder-7.2 inches in dia.
0M HNO_3, 450 g/l Pu Slab -3.8 inches thick
3. FIXED POISONS	◦ Permissible, provided neutron absorption is sufficient to prevent criticality under all foreseeable process conditions. ◦ Annual inspections required to determine integrity of poison.
4. ADMINISTRATIVE
CONTROLS	◦ Should be supplemented by instruments or mechanical devices whenever possible; ◦ Multiplication Factor (K_{eff}) must be no greater than: 0.98 - System well-defined and reliable experimental data exist for closely similar systems; 0.95 Limited experimental data exist and relatively large extrapolations are necessary; and 0.90 No experimental data are available and calculations must be based on theory.

1. Geometrically Safe Equipment - equipment that has been designed to be subcritical by virtue of neutron leakage under all possible conditions of inventory and reflection. Equipment of the required size (a cylinder 1.7 inches in diameter or a slab 0.25 inches thick in the case of plutonium-239) however, would be impractical and prohibitively expensive in a large separations plant.
2. Geometrically Favorable Equipment - equipment that is subcritical by virtue of neutron leakage under the worst foreseeable process conditions. To be considered *geometrically favorable*, the diameter of a cylinder, or thickness of a slab should be no greater than 85 percent of the minimum value that can be calculated to permit criticality under the worst foreseeable conditions. For containers of unspecified or irregular shape, the permissible volume is no greater than 75 percent of the minimum volume that would be critical at optimum concentration.
3. Fixed Poisons - the use of fixed poisons is permissible, provided the equipment is so constructed that neutron absorbers in the structure prevent criticality under all foreseeable process conditions. While sufficient poison material theoretically can be placed in a system to assure criticality prevention, it may not be practical to insert a sufficient quantity of poison to achieve this condition. Fixed poisons are therefore used in such a manner as to permit a predetermined incremental increase in the system geometry, the allowable mass or concentration, or a combination of the three. The use of a fixed poison in the Hanford Purex Plant requires the scheduling of annual inspections to determine the integrity of the poison.
4. Administrative Controls - included in this general classification are mass and concentration controls, array limits and the use of soluble poisons. Administrative controls are permissible when the use of favorable geometries or fixed poisons is not practical to assure criticality prevention. When administrative controls are used, however, additional restrictions should be incorporated into process conditions and operating procedures to insure criticality prevention even though a single credible error or omission has been committed. In addition, instruments and/or mechanical devices should be provided to assist in the measurement and control of the process within the prescribed limits.

The definition of safe administrative control limits involves the application of technical criteria which are beyond the scope of this paper. However, general requirements and limit values have been established at Hanford for application to chemical processing plants. The values presented in Table II have been established as permissible upper limits for each mechanism of control.

TABLE II

ADMINISTRATIVE CONTROL CRITERIA AND LIMITS

GENERAL REQUIREMENTS

- Maximum permissible quantity or concentration must be less than 50 percent (75 percent behind massive shielding) of that required to sustain a nuclear chain reaction under any credible conditions.
- The limit specified for a system shall be such that the value of K_{eff} at the allowable concentration (or a double-batch condition) shall not exceed the applicable value shown in Table I.

CONTROL LIMITS

- Mass Control 120 grams ^{239}Pu /Sq. Ft.* (Behind shielding, 180 g/sq. ft.)
- Concentration Control-Solutions 4 g Pu/l (Behind shielding, 6 g/l)
- Concentration Control-Arrays Dispersion in space must be such that the reactivity of a single container is not significantly increased by interaction with adjacent units. Container Separation Limit: 18 inches Ctr-to-Ctr, min.
- Soluble Poisons Must be in sufficient concentration to prevent criticality under all foreseeable process conditions.

Use as a primary control is not permitted in an unshielded facility.

*Bottom horizontal cross-section of vessel.

- a. Control by Mass Limits - the quantity of fissile material in a given location is limited to less than half of that required for criticality under any credible conditions.
- b. Control by Concentration Limits - Solutions - the concentration of fissile material dissolved or dispersed in another medium should be limited so that neutron absorption in the diluent prevents criticality. One of our maximum limits is six grams of plutonium-239 per liter of solution. Another concentration limit that is useful prior to partitioning of uranium and plutonium is expressed in terms of grams of plutonium per ton of uranium. The plutonium gram per ton limit is dependent, of course, on the ^{235}U content of the irradiated uranium.
- c. Control by Concentration Limits - Arrays - the dispersal in space of discrete accumulations or containers of fissile material can be controlled with respect to geometry and distance so that a single, subcritical unit is effectively isolated from all adjacent units. A standard minimum center-to-center distance frequently used at Hanford for small (geometrically favorable) cylinders is 18 inches.
- d. Control by Soluble Poisons - The use of neutron-absorbing materials (such as boron or cadmium) is permissible, provided they are in solution with the fissile materials in sufficient concentration to prevent criticality under all foreseeable process conditions. Other restrictions applying to the use of soluble poisons include consideration of mechanisms that might change the concentration of the poison, as well as the concentration of fissile material. The use of soluble poisons as a primary means of precluding criticality is not permitted in an unshielded facility.

PHILOSOPHY AND BASES FOR APPLYING CRITICALITY CONTROLS

The methods used for defining the minimum conditions for criticality have been greatly refined in recent years. A number of computer programs have been developed and are very useful in calculating the approach to criticality for well-defined systems. In addition, facilities such as the Critical Mass Laboratory at

Hanford, are now available for verifying experimentally, the computer data. Considerable uncertainty is still involved in many operational systems, however, because of the extreme difficulty in predicting the exact chemical composition of the process solutions and the conditions affecting neutron moderation and absorption. In addition, regardless of the refinement introduced by computer programs and experimental facilities, criticality data must be extrapolated from measurements of sub-critical systems.

The uncertainties involved in criticality measurements and calculations and in predicting process conditions, together with the human factor (and the concomitant probability of error in plant operation and control) necessitate a conservative approach in specifying standards and limits for criticality prevention.

(This does not mean; however, that the restrictions and limits are necessarily unchangeable. To the contrary, as experimental work and proven calculation methods more accurately define the conditions of criticality for various systems, less restrictive controls and limits generally result.)

This generally conservative philosophy is implemented in the nuclear safety practices at the Hanford chemical processing plants primarily by the inclusion of safety factors in limit calculations (to allow for uncertainties) and the application of a *multiple error* philosophy in establishing nuclear safety specifications and operating requirements.

The following are examples of the application of safety factors in the specification of nuclear safety limits.

- a. The poisoning effect of nitrate ion in Purex process solutions is not normally taken into consideration.

- b. The full ^{235}U burnup during irradiation of uranium fuels is not normally considered when specifying the safe concentration of fissile material in uranium-plutonium solutions.

It is emphasized that the use of the above safety factors is not required by the criticality prevention criteria. The full benefit of poisons or burnup may be utilized, provided there is positive knowledge and control of the actual conditions.

The *multiple error* philosophy, as applied in establishing nuclear safety limits, requires that conditions and controls be maintained such that at least two* simultaneous and independent operational errors, equipment failures, or process deviations are necessary for a nuclear excursion to occur. In the case of a continuous process, for example, this would require exceeding the concentration limit plus complete precipitation or denitration. For a batch process, fissile material mass limits would be such that merely *double-batching* would not be sufficient to initiate a nuclear excursion.

APPLICATION

Consideration of criticality prevention principles, and preparation of the criticality safety analysis, should logically commence with flowsheet design and continue through equipment and facility design. Finally, safety considerations for personnel, plant and environment require that criticality prevention principles and philosophy become an integral part of the process and operational controls, techniques, and procedures.

FLOWSHEET DESIGN

Consideration of criticality prevention principles during flowsheet definition can greatly simplify their application during plant

*At least two errors are required in remote, heavily-shielded processing areas. Generally, three errors are required in contact-operated areas.

operation. For example, the recycle of fissile material via waste streams, as well as internal reflux in solvent extraction equipment should be taken into account in defining fissile concentrations in feed streams. Fissile concentrations and masses must be considered, in relation to the maximum safe values, for any streams or solutions subject to concentration. The possibility of precipitation must be considered unless methods are incorporated into the design to positively prevent the accidental addition of precipitating agents to the system. The possibility of concentration of fissile material to an unsafe level in the organic solvent phase, while not a problem in the standard Purex flowsheet, must be given special attention in the definition of enriched uranium or thorium- ^{233}U process flowsheets.

FACILITY DESIGN

In the design of a nuclear fuels processing facility, consideration should be given, where appropriate, to the use of geometrically favorable equipment. As mentioned previously, this concept not only applies to vessel shape and size, but to its interaction with other vessels and piping, moderating effects of construction materials, and possible effects of pressurizations, corrosion, or other incidents on the vessel integrity. For instance, a thin slab shape is a useful design for a geometrically favorable tank. Very strong internal bracing is required, however, to assure that the critical thickness dimension is maintained. Without strong bracing, slight pressure changes - on the order of a few inches of water - can cause intolerable changes in the tank thickness.

Other shapes have been used successfully at Hanford, including cylinders (seven inches or less in diameter) multiple-cylinder *bank* tanks and annular vessels. Both flat slabs and annular cylinders, for example, have been used for phase disengaging sections on the pulse columns. (Annular disengaging sections are currently in favor, because they are easier to equip with interface control devices.) Multiple-cylinder *bank* tanks are convenient fissile solution storage vessels and are relatively inexpensive. Mixing is a problem in bank tanks, but can be accomplished by external circulation or by controlled agitation.

Another area that should be given criticality prevention consideration in facility design is the drainage collection sump system in the processing cells and the possibility of fissile material leaking into the sumps. Product rework capability could be severely restricted if the sumps are not excluded or designed to be geometrically favorable.

The use of a fixed poison is an acceptable alternative to equipment of geometrically favorable design. The use of fixed poison, however, may introduce complications later, during normal operation, because its safety is contingent on it retaining its integrity and position in the equipment. Periodic inspections of a fixed poison should be made to insure its continued effectiveness. As mentioned previously, the required frequency at Hanford is at least once per year. Since inspections of remote equipment are very difficult, fixed poisons have rather limited applicability in practice.

Finally, if the use of either geometrically favorable equipment or fixed poisons is impractical, administrative controls must be relied upon for criticality prevention. Complete coordination between the design engineer and the process and manufacturing engineers in considering nuclear safety control methods can greatly increase the reliability of administrative controls. Fissile material monitors (either neutron counters or alpha scintillation counters) are very helpful aids to criticality prevention.

Another way in which the design engineer can be of great assistance in improving administrative control is in the design of appropriate interlocks, sequencing switches, and locking gang valve switches to prevent the accidental addition of a precipitating agent to a solution of fissile material.

APPLICATION TO PROCESSING OPERATIONS

The preparation of the criticality safety analysis and specifications, while of major importance, is only the first step in the plant application of criticality safety principles. Practical controls and procedures must then be established to assure that the processing operations are conducted within the prescribed parameters. A convenient mechanism for accomplishing this is the Standard Operating Procedure. The Standard Operating Procedures should clearly describe the operation to be carried out, special safety considerations, and the actions necessary to maintain control within the specified limits. The procedures should logically be prepared and reviewed by persons technically knowledgeable of the process and operation to be covered. Approval should be by persons responsible for the process control and plant operations functions.

Standard Operating Procedures generally prescribe methods and techniques for use by personnel in operating mechanical equipment. Other measurements and control criteria are necessary for monitoring of the process for conformance to flowsheet and specification limits. Chemical analysis of selected process stream samples backed up by process instrumentation, provides an acceptable measurement method. The samples should be taken on a routine, prescribed (and documented) frequency schedule to assure consistency. The analytical schedule should also be shown on the sample schedule. Those samples and analyses and the minimum safe frequency required for criticality prevention should be clearly designated. Any deviation from this minimum schedule should be compensated for either by sampling of alternate streams or by supplementing other measurement and control methods.

The criticality prevention specifications are designed to minimize the probability of criticality by establishing maximum safe limits or restrictions for processing operations. Thus, violations of the specified limits and restrictions have serious implications and require formal investigation. More restrictive limits are frequently established for process control standards. (These standards, however, are generally well above routine processing levels.) The sample analyses are compared to these standards and adjustments made or responsive action taken to maintain the process within the limits designated in the standards.

Instrumentation, in addition to being absolutely essential for operational control of remote equipment, is a valuable aid in criticality prevention. Neutron monitors located in the process cells and alpha scintillation monitors located at the sample retrieval stations provide continuous measurements of the fissile content of selected, sensitive vessels and streams. Neutron and alpha scintillation monitors have proved to be very valuable criticality prevention tools, particularly around the uranium-plutonium partitioning system.

Other, more common instrumentation is also heavily depended upon for criticality prevention. In processing areas where concentration or liquid level measurements are sensitive, such as the final product treatment and loadout facilities, weight factor and specific gravity instruments and both audible and visual alarms are important. Instruments in this service must be kept in good repair and checked for proper calibration on a regular schedule.

All of the factors discussed above--process and equipment design, chemical analysis, and instrumentation--are important considerations for nuclear criticality prevention. Perhaps the single most important factor - indeed, an absolutely essential factor in any nuclear criticality prevention program, however, is the attitude of the operating personnel regarding nuclear safety. The process operators must be made aware of the serious consequences of off-standard operations and their complete cooperation in reporting and correcting such conditions is essential to effective administrative control for criticality safety. This necessary concern for criticality prevention can be obtained only by effective and continuing communications from management through line supervision to the operating personnel.

SUMMARY

A primary concern in the design and operation of a nuclear fuels processing plant is the prevention of an uncontrolled nuclear chain reaction, or nuclear criticality excursion. Effective plant application of criticality prevention principles should commence with process flowsheet design and continue through equipment and facility design and construction. Finally, safe and successful plant operation requires that criticality prevention principles and practices be accorded paramount consideration by all members of the production and support teams.